

SPECTROSCOPIC MANIFESTATION OF THE UNUSUAL PROPERTIES OF STRONG NUCLEAR INTERACTION

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Abstract

The dependence of the proton - neutron interaction in the deuterium nucleus on the distance between nucleons which was not available on accelerators hence it was first - time measured. Experimentally, neutron electron binding energy equal to 106 meV was found in diamonds and LiH which is 128 times less than proton - electron coupling energy (13.6 eV) and its value is perfectly agreed with Breit's theory. The results of our experiments make it possible to determine not only the constant of strong nuclear interaction equal 2.4680, but also to predict the mass of the boson carrying the interaction between neutron and electron equal 3.5 keV. Moreover, the creation of mass in massless Dirac fermions (leptons) in graphene by strong nuclear interaction has been demonstrated for the first time.

Keywords: strong interaction, hadrons, quarks, gluons, excitons, phonons, quantum electrodynamics, quantum chromodynamics

I. Introduction.

Simple atoms, like hydrogen, being essentially quantum electrodynamics (QED) systems, allow highly accurate theoretical predictions. QED is the first successful and still the most successful quantum field theory. The spectacular success of QED gives physicists great confidence in Maxwell's equation on the one hand and Dirac's equation on the other, yet something is missing in relations between them (see, e.g. [1]). After the discovery of the neutron in 1932 by Chadwick, there was no longer doubt that the building block of nuclei are proton and neutron (collectively called nucleons). The discovery of the neutron may be viewed as the birth of the strong nuclear interaction: it indicated that the nuclei consists of protons and neutrons and hence the presence of a force that holds them together, strong enough to counteract the electromagnetic repulsion. Theory strong nuclear interaction is the heart of quantum chromodynamics (QCD) which is part of the Standard Model (SM) [2, 3], therefore the base exchange is the gluon which mediates the forces between quarks. Nucleus is a bound system of strongly interacting protons and neutrons (more generally baryons and mesons - this is the reason for the collective name hadrons) holds atomic nuclei together and, in another context, binds quarks within hadrons. The baryons are bound states of the three quarks and mesons are composed of quark and antiquark (see, e. g. [4]).

The subject of elementary particle physics to have begun with the discovery of the electron more than century ago. In the following 50 years, one new particle after another was discovered, mostly as a result of experiments with cosmic rays, the only source of very high energy particle then available. After second war the accelerator technique was used to study the elementary particle physics. Nuclear and particle physics are essentially at the forefront of nowadays understanding of physics. Nucleus is a bound system of strongly interacting protons and neutrons. Unfortunately, modern

Quantum Electrodynamics (QED) does not provide us with the tools to calculate the bound states properties of the proton (deuteron) from first principles [1]. The present review is devoted to the results of measurements of residual strong nuclear interaction via the study the low - temperature optical spectra (reflection, photoluminescence) of the LiH (without strong interaction in hydrogen nucleus) and LiD (with strong interaction in deuterium nucleus) crystals which are different by term of one neutron from each other as well as very rich of diamond allotropes (diamond, graphite, graphene, fullerene). We should repeat that nowadays in text books and elsewhere the separation of electromagnetic and strong interaction is tacitly assumed. It is very strange because up to present time we do not even know the strong nuclear force very well. The origin of the strong interaction is very important especially in the problem of all force unification (see, also [5]). The results of the paper [6] have shown a new light on some residual strong nuclear interaction (ultimately based in the character of magnetic forces, the electromagnetic or color origin, which by their very nature, are difficult to conceal within the elusive nucleon physical boundary) between both kind of forces which experimentally manifested through isotopic shift of zero phonon line in optical spectra.

As indicated above, there is a common place in Standard Model (SM) of modern physics; that the strong force does not act on leptons (see, also [5]). Following this conclusion we do not must to observe the dependence of the optical properties of solids on addition neutrons in substance. This contradict the history of the development of isotope effect. The first attempt to discover an interaction between neutrons and electrons was made by Dee [7] in the same year, 1932 in which the neutron was discovered by Chadwick [8]. In 1936, Condon [10] pointed out that the existence of a neutron - electron interaction would give rise to an isotope shift in spectral lines of atoms, which was observed for the first time by Aronberg in 1918 [9] in the line spectra of Pb isotopes (see, also review [11]). The existence of a weak attractive interaction between electrons and neutrons has been described in the series papers by Foldy [12]. Foldy has showed that neutron - electron interaction has two contributions - one arising from anomalous magnetic moment of the neutron (see, also [5]) and the other from an intrinsic Darwin coefficient [13] - electric field. This picture was justifiable in meson theory though both of these contributions have a common origin [12, 14]. In all methods of the measurements (see, e.g. [14]) there are principal troubles connected with the necessity of introducing large corrections in size of order of the investigated effect of neutron - electron interaction. Besides there is intrigue in the fact that all known experimental values (see [15] and references quoted therein) were scattered around the so called Foldy scattering length in the interval $\pm$

10%. The main conclusion of the fundamental papers by Foldy is that the intrinsic neutron - electron interaction is essentially an electromagnetic interaction between the neutron and the charge density producing an external electromagnetic field by electron [12, 14].

According to contemporary physics the strong force does not act on lepton (electrons, positrons, muons and neutrinos), but only on protons and neutrons (more generally, on baryons and mesons - this is the reason for the collective name hadrons) (see, however below). It should be add that the forces between the quarks must be long range, because the gluons (as photons) have zero mass. This does not imply that the forces between hadrons are also long - range, because hadrons have zero color charges overall. The forces between the colorless hadrons are the residues of the for as between their quark constituents, and cancel when the hadrons are far apart [2 - 4]. In 1935, Yukawa [16] pointed out that the nuclear force could be generated by the exchange of a hypothetical spinless particle, provided its mass intermediate between the masses of proton and electron - a meson. Yukawa predicted the pion [1 - 4]. The macroscopic manifestations of the strong interaction are restricted up to now to radioactivity and the release of nuclear energy. Yutaka's potential has the form

$$V = g \frac{e^{-kr}}{r^n}, \quad (1)$$

for some n . Here the strength of the force is measured by the constant k (see, also [5]). The most important feature Yukawa's forces is that they have a small range ($\sim 10^{-15}$ m). The central dogma of atomic physics after Yukawa's paper that proton - electron attraction could be explained in terms of classical electrostatic theory, while the strong force effects were essentially new and inexplicable. So, far the best theoretical guess is the Yutaka potential, but it is a static potential not dependent on velocities of the nucleons. A static force is not a complete one because it can not explain the propagation of the nuclear interaction. Moreover, as was indicated above, a phenomenological Yukawa potential can not be directly verified experimentally. We should note that nowadays in text books and elsewhere the separation of electromagnetic and strong interaction tacitly assumed. It is very strange up to present time we do not even know the strong force very well. And what is more we have some contradiction taking into account that the forces between quarks must be long - range, because the gluons have zero mass. But as was mentioned above the force between colorless hadrons is short - range, when the distance between hadrons is more than nuclear size [1]. In connection of our brief analysis, we must emphasize that outanding works on hadrons mechanics [17] deserve separate and fundamental

study.

One may consider four families of particles in order of increasing rest mass; the first contains only one member, the proton, a boson of spin 1. The second family, leptons, contains fermions of spin 1/2, lighter than proton. Leptons are subject to electromagnetic and Fermi interaction only, not the strong interaction (see however below). The third family, mesons, comprises bosons of spin 0. These are heavier than the leptons, lighter than the protons, and the subject to all three types of interactions; weak, strong and electromagnetic. The fourth family, baryons, comprises the proton and heavier fermions. Baryons are subject to all three types of interactions; those heavier than neutron are called hyperons.

Table 1. The four fundamental forces.

Interaction	FQ	Mass	Range (m)	RS	Spin	TC-S (m ²)
TTS(s)						
Str	Gluon	0	10 ⁻¹⁵	1	1	10 ⁻³⁰
10 ⁻²³						
We	W , Z	81, 93 GeV/c ²	10 ⁻¹⁸	10 ⁻⁵	1, 1	10 ⁻⁴⁴
10 ⁻⁸						
Elec	Photon	0	∞	1/137	1	10 ⁻³³
10 ⁻²⁰						
Gra	Graviton	0	∞	10 ⁻³⁸	2	-
Here - FQ field quant, RS relative strength, TC - S Typical cross - section, TTS - Typical time scale.						

The Table 1 (see also [19]) given for the strength and range of the forces come from a comparison of the effects they produce on two protons. In some respect these resemble an ordinary Newtonian force between the protons, varying with the distance between them as if the force was derived from a potential function (1): for some n. This is an inverse - power force which is diminished by an exponential factor at distances larger than a certain distance R, the range of the force. The strength of the force is measured by the constant k. The unit of strength is $hc/2\pi$ where h is Planck's constant and c the speed of light. Thus nuclear physics was essentially the paradigmatic example of understanding particle physics. The modern quantummechanical view of the three fundamental forces (all except gravity) is that

particles of matter (fermions neutrons, protons, electrons) do not directly interact with each other, but rather carry a charge, and exchange virtual particles (gauge bosons photons, gluons, gravitons) which are the interaction carriers or force mediators. As can be seen from Table 1, photons are the mediators of the interaction of electric charges (protons, electrons, positrons); and gluons are the mediators of the interaction of color charges (quarks). In our days, the accepted view is that all matter is made of quarks and leptons. As can be seen, of the three pairs of quarks and leptons, one pair of each - the quark u and d and the leptons e and ν_e (electrons neutrino) - are necessary to make up the every day world, and a world which contained only these would seem to be quite possible.

As we can see above the method of the scattering particles allows to determine only length of scattering as well as the size and depth (highest) of the neutron's potential. In the present paper we attempt to measure the energy of the neutron - electron interaction via spectroscopic study of the optical characteristics of solids with isotope effect at low temperature. Therefore, it is purpose of our paper to advance a description of the manifestation of strong nuclear interaction in solids, using partly published and new non - accelerator experimental results. Our spectroscopic measurement of the low temperature optical characteristics of $\text{LiH}_x\text{D}_{1-x}$ crystals is permitted to quantitative study of the dependence of strong coupling constant, α_s , on the proton - neutron distance in the deuteron nuclei. Carbon, being the second element after silicon on Earth, has unusually wide range of allotropic compounds with completely different physical properties [20]. The use of our object allows the investigation of not only the isotope effects in lattice dynamics (vibrational, elastic and thermal properties), but also the influence of such effects on the electronic excitations (for example, the renormalization of the band - to - band transition energy E_g). We emphasize that the study of the isotope effect remains at the forefront not only in materials science, but also nuclear and high energy physics.

Isotopically pure materials represent a separate class of modern materials science, which is an accelerator of the development of the latest fine technology. First of all, it is necessary to remember due to this explosive leap in the development of electronic and optical technologies. It is thanks to isotopically pure silicon used in personal computer processors that it has been possible to step beyond the 3GHz threshold. Moreover, isotopically pure germanium is the best weak signal detector. Low dimensional structures of quantum wires (QW), quantum dots (QD) received a new impetus for development when using isotopically pure materials, as well as artificially regulating the isotopic composition of a material. We emphasize that due to the wide class of allotropic carbon compounds, the physical properties of which vary from

transparent in the ultraviolet (UV) range (diamond) to opaque (graphite, graphene), from dielectric (diamond) to a compound with metallic conductivity (graphene) and also which have extremely different elastic and thermal properties - all of the above mentioned characteristics exhibit isotopic dependence. We must substitute ^{12}C for a heavy isotope ^{13}C . Semiconductor graphene could come to replace all silicon electronics. Of course, the new technology for the development of quantum computer (QC) processors is based on the use of materials with different spin values. Great demand for isotopically pure materials from control agencies. For example, a special requirement for isotopically pure silicon, from which it is planned to make a control sample of 1 kg. We will not at all touch upon the very wide applications of pure isotopes in biophysics and medicine. As an example, let us point out that only one country (USA) uses tens of thousands tons of different isotopes per year.

The present paper reviews of recent research work on the preparation and remarkable properties of isotope - mixed systems, covering wide range of topics from physics of elementary particles, materials science to engineering applications, including in quantum information and quantum computers. Artificial activation of the strong interaction by adding of one neutron to the nucleus causes the global reconstruction of the macroscopic characteristics of solids. The experimental evidence of macroscopic manifestation of the strong interaction in optical spectra of solids which are different by term of one neutron from each other (using LiD crystals instead LiH) has been presented. The initial components of mixed crystals $\text{LiH}_x\text{D}_{1-x}$ are LiH (no strong interaction in the hydrogen nucleus) and LiD (with strong interaction in the deuterium nucleus) crystals are dielectrics with cubic symmetry. The change of the concentration of deuterium in mixed crystals $\text{LiH}_x\text{D}_{1-x}$ determines the change in the distance between nucleons, which is directly manifested in the position of the phononless free exciton emission line in the low - temperature luminescence spectrum. A series of such measurements made it possible to construct a function of the dependence of the energy of strong nuclear interaction on the distance between nucleons in the deuterium nucleus. This evidence is directly seen from luminescence (reflection) and scattering spectra. As far as the gravitation, electromagnetic and weak interactions are the same in both of kind crystals, it only emerges the strong interaction in deuterium nucleus. Therefore a sole conclusion is made that the renormalization of the energy of electromagnetic excitations (electrons, excitons, phonons) is carried out by the strong nuclear interaction. The measurements of the dependence of the energy of strong nuclear interaction on the distance between nucleons in a nucleus were performed for the first time and this results allow to find the maximum possible value of $\alpha_s = 2.4680$. Our non - accelerator results opens new

avenue in the investigation of the propagation the force of strong nuclear interaction in the wide value range by means the condensed matter alike traditional accelerator methods. Carbon, being the second element after silicon on Earth, has unusually wide range of allotropic compounds with completely different physical properties. The use of our object allows the investigation of not only the isotope effects in lattice dynamics (vibrational, elastic and thermal properties), but also the influence of such effects on the electronic excitations (for example, the renormalization of the band - to - band transition energy E_g). We emphasize that the study of the isotope effect remains at the forefront not only in materials science, but also nuclear and high energy physics.

II. Experimental.

The apparatus used in our experiments has been described in several previous publications [21 - 23]. For clarity, we should mentioned here that immersion home - made helium cryostat and two identical double - prism monochromators were used. One monochromator was used for the excitation and the other, which was placed at right - angle to the first for analyzing the luminescence and scattering of light. In our experiments we investigated two kinds of crystals (LiH and LiD) which are differ by a term of one neutron. Lithium hydride and lithium deuteride are ionic insulating crystals with simple electronic structure, four electrons per unit cell, both fairly well - described structurally (neutron diffraction) and dynamically (second - order Raman spectroscopy) and through ab initio electronic structure simulation. Among other arguments, LiH and LiD are very interesting systems due to their extremely simple electronic and energy structure and to the large isotopic effects when the hydrogen ions are replaced by the deuterium ones. On the other hand, the light mass of the ions, specially H and D, makes that these solids have to be considered like quantum crystals, and consequently, described theoretically by quantum theory. In a solid one deals with a large number of interacting particles, and consequently the problem of calculating the electronic wave functions and energy levels is extremely complicated. It is necessary to introduce a number of simplifying assumptions. In the first place we shall assume that nuclei in the crystalline solid are at rest. In an actual crystal this is of course never the case, but the influence of nuclear motion on the behavior of electrons may be treated as a perturbation for the case in which they are assumed to be at rest. Even with above assumption, however, we are still with a many - electron problem which can be solved only be approximative methods. In the case of solids, the most

important approximative method which has been applied extensively is the so – called one – electron approximation. In this approximation the total wave function for the system is given by a combination of wave functions, each of which involves the coordinates of only one electron. In other words, the field seen by a given electron is assumed to be that of the fixed nuclei plus some average field produced by the charge distribution of all other electrons.

The difference between a good conductor and a good insulator is striking. The electrical resistivity of a pure metal may be as low as 10^{-10} ohm-cm at a temperature of 1 K, apart from the possibility of superconductivity. The resistivity of good insulator may as high as 10^{22} ohm-cm. To understand the difference between insulators and conductors, we shall use the band – gap picture (Fig. 2 below). The possibility of band gap is the most important property of solids.

The single crystals of LiH and LiD were grown from the melt by the modified method of Bridgeman - Stockbarger (see [24, 25]). The crystals were synthesized from ^7Li metal and hydrogen of 99.7% purity and deuterium of 99.5% purity. Virgin crystals had a slightly blue - grey color, which can be attributed to nonstoichiometric excess of lithium present during the growth cycle. On annealing for several days (up to 20) at 500°C under ~ 3 atm of hydrogen or deuterium, this color could be almost completely eliminated. Because of the high reactivity and high hygroscopy of investigated crystals should be protected against the surrounding atmosphere. Taking into account this circumstance, we have developed special equipment which is allowed to prepare samples with a clean surface cleaving them in the bath of helium cryostat with normal or superfluid liquid helium [22]. The samples with such surface allow to perform measurements during 15 hours. An improved of LiH (LiD) as well as mixed crystals were used in the present study. In spite of the identical structure of all free - exciton luminescence spectra, it is necessary to note a rather big variation of the luminescence intensity of the crystals from the different batches observed in experiment. Experimental set up for diamond and graphene is described in relatively cited below papers. The crystal periodicity leads to the formation of a band energy structure (Fig. 1). An electron from valence band is excited into conduction band.

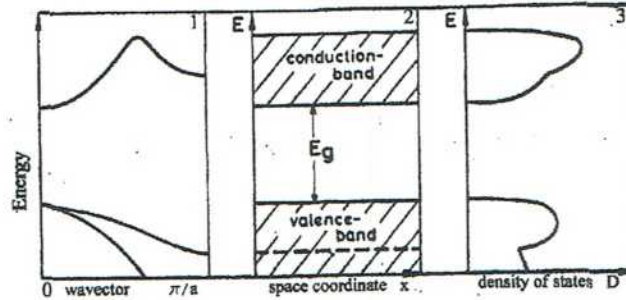


Fig. 1. Various possibilities to present the band - structure of homogeneous undoped insulator (semiconductor). 1 - the dispersion relation, i.e. the energy E as a function of the wave vector $\vec{k}$, 2 - the energy regions of allowed and forbidden states as function of a space coordinate x and 3 - the density of states (all curves are schematic ones) [22].

The attractive Coulomb potential between the missing electron in the valence band, which can be regarded as a positively charged hole, and the electron in the conduction band gives a hydrogen - like spectrum with an infinite number of bound state and ionization continuum Fig. 2. Below we will briefly describe the results of the optical spectroscopy of isotope - mixed solids. For this purpose we use crystals LiH (without strong interaction in the nucleus) and LiD (with strong interaction in the deuterium nucleus) which differ by term of one neutron from each other as well as diamond with different concentration of different isotopes.

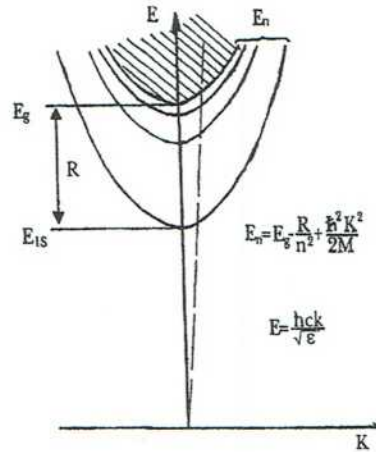


Fig. 2. Discrete and continous (hatched area) Wannier - Mott exciton energy spectrum taking into account its kinetic energy $\hbar^2 k^2 / 2M$. The broken line connects to the dispersion of light in the medium. R - is the exciton binding energy, $n = 1, 2, 3, \dots$

As demonstrated early (see, e.g book [24]) most low - energy electron excitation in LiH (LiD) crystals are the large radius excitons [24, 26].

III. Results.

Photoluminescence is the optical radiation emitted by a physical system (in excess of the thermal equilibrium blackbody radiation) resulting from excitation to a nonequilibrium state by irradiation with light. Photoluminescence is also rapidly evolving into major basic research tool comparable to absorption (reflection) measurements in importance. Two reason for this stand out as significant. First is the sensitivity of the luminescence technique. It often happens that features which are just discernible in absorption will completely dominate the luminescence spectra. The converse is also sometimes true, making luminescence and absorption (reflection)

complementary techniques. Second is the simplicity of data collection. In last half-century the luminescence method has become one of the most common techniques for studying excitons in dielectrics and semiconductors. While the structure of spectra of fundamental reflection (absorption) depends on the internal degrees of freedom of Wannier – Mott exciton, the structure and shape of the luminescence spectrum are determined primarily by its external degrees of freedom. The latter are associated with the translation motion of large – radius exciton as a whole, with the translation mass $M = m_e + m_h$, where m_e and m_h – effective masses of electron and hole, respectively.

Free exciton luminescence is observed when LiH (LiD) crystals are excited in the midst of the fundamental absorption. The spectrum of free exciton photoluminescence of LiH crystals cleaved in superfluid helium consists of a narrow (in the best crystals, its half - width is $\Delta E \leq 10$ meV [27]) phononless emission line and its broader phonon repetitions, which arise due to radiated annihilation of excitons with the production of one to five longitudinal optical (LO) phonons (see Fig. 3).

The phononless emission line coincides in an almost resonant way with the reflection line of the exciton ground state which is indication of the direct electron transition $X_1 - X_4$ of the first Brillouin zone [28]. The lines of phonon replicas form an equidistant series biased toward lower energies from the resonance emission line of excitons. The energy difference between these lines in LiH crystals is about 140 meV, which is very close to the calculated energy of the LO phonon in the middle of the Brillouin zone [29] and which was measured in (see, e.g. [24] and references quoted therein). The isotopic shift of the zero - phonon emission line of LiH crystals equals 103 meV. As we can see from Fig. 3 the photoluminescence spectrum of LiD crystals is largely similar to the spectrum of intrinsic luminescence of LiH crystals. There are, however, some distinctions one is related. Firstly the zero - phonon emission line of free excitons in LiD crystals shifts to the short - wavelength side on 103 meV. These results directly show the violation of the strong conclusion (see, e.g. [1 - 4]) that the strong force does not act on leptons. The second difference concludes in less value of the LO phonon energy, which is equal to 104 meV. The simplest approximation, in which crystals of mixed isotopic composition are treated as crystals of identical atoms having the average isotopic mass, is referred to as virtual crystal approximation (VCA) [24].

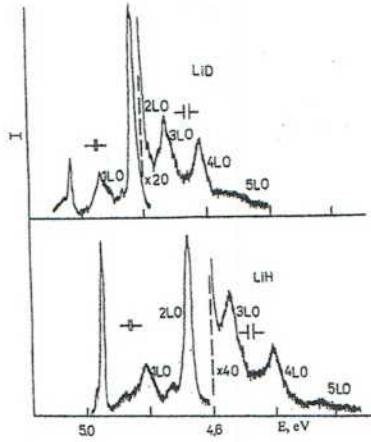


Fig. 3. Photoluminescence spectra of free excitons at 2 K in LiH and LiD crystals cleaved in superfluid helium.

When light is excited by photons in a region of fundamental absorption in mixed $\text{LiH}_x\text{D}_{1-x}$ crystals at low temperature, lines luminescence of free excitons are observed (Fig. 4), like in the pure LiH and LiD crystals. As before [24], the luminescence spectrum of crystals cleaved in superfluid liquid helium consists of the relatively zero - phonon line and its wide LO replicas. For the sake of convenience, and without scarfing generality, Fig. 4 shows the lines of two replicas. Usually up to five LO repetitions are observed in the luminescence spectrum as described in detail in [30]. In Fig. 4 we see immediately that the structure of all three spectra is the same. The difference is in the distance between the observed lines, as well as in the energy at which the luminescence spectrum begins, and in the half - width of the lines.

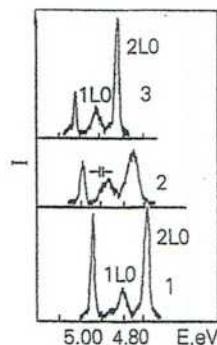


Fig. 4. Photoluminescence spectra of free excitons in LiH (1), $\text{LiH}_x\text{D}_{1-x}$ (2) and LiD (3) crystals cleaved in superfluid helium at 2 K. Spectrometer resolution is shown.

The change of the concentration of deuterium in mixed crystals $\text{LiH}_x\text{D}_{1-x}$ determines the change in the distance between nucleons, which is directly manifested in the position of the phononless free exciton emission line in the low - temperature luminescence spectrum. A series of such measurements made it possible to construct a function of the dependence of the energy of strong nuclear interaction on the distance between nucleons in the deuterium nucleus. This evidence is directly seen from luminescence (reflection [24]) and scattering spectra [30]. The X - ray diffraction investigations show that the $\text{LiH}_x\text{D}_{1-x}$ mixed crystals [31] form a continuous row of the solid solution and testify themselves like a virtual crystal with a variable lattice constant (and exciton radius, respectively) that obeys Vegard's law (see Fig. 5)

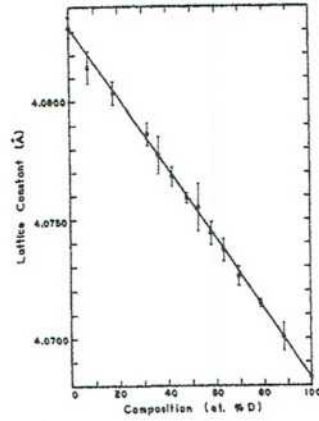


Fig. 5. Lattice constant in the ${}^7\text{Li}(\text{H,D})$ crystals plotted against the isotopic composition [31].

Fig. 6 shows the concentration dependence of the energy of interband transition E_g (see Fig. 1). As can be seen from Fig. 6, VCA method (the straight dashed line) cannot describe observed experimental results. This dependence has a nonlinear character.

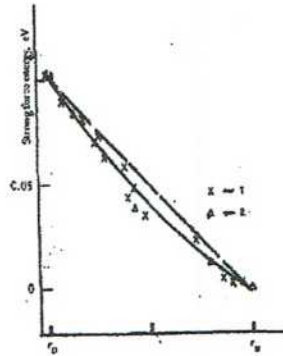


Fig. 6. Dependence of the interband transition (strong interaction) energy E_g in mixed crystals $\text{LiH}_x\text{D}_{1-x}$ on the concentration x (number of neutrons). Here r_D and r_H are the radii of deuterium and hydrogen nuclei according CODATA recommended values. The straight line is the linear dependence $E_g = f(x)$ in the VCA model. The solid line corresponds to calculation using the polynomial of second degree [22].

Comparison the experimental results on the luminescence (reflection) and light scattering in the crystals $\text{LiH}_x\text{D}_{1-x}$ which differ by a term of one neutron only is allowed to the next conclusions;

1. At the adding one neutron (using LiD crystals instead LiH ones) is involved the increase exciton energy on 103 meV.
2. At the addition one neutron the energy of LO phonons is decreased on the 36 meV, that is direct seen from luminescence and scattering spectra.

Both characteristics are macroscopic.

Another very interesting example is carbon. Carbon atom is built from 6 protons, A neutrons and 6 electrons, where $A = 6$ or 7 , yield the stable isotopes ^{12}C and ^{13}C , respectively, and $A = 8$ characterizes the radioactive isotope ^{14}C [32]. The isotope ^{12}C , with nuclear spin $I = 0$, is the most common one in nature with 99% of all carbon atoms, whereas only $\approx 1\%$ are ^{13}C with nuclear spin $I = 1/2$. There only traces of ^{14}C (10^{-12} of all carbon atoms) which transforms into nitrogen ^{14}N by β - decays [33]. Although ^{14}C only occurs rarely, it is important isotope used for historical dating (see,

e.g. [37]). Carbon, one of the most elements in nature, still gives a lot surprises. It is found in many different forms - allotropes - from zero dimensional fullerene, one dimensional carbon nanotubes, two dimensional graphene and graphite, to three dimensional diamond - and the properties of the various carbon allotropes can vary widely [20, 34]. Fullerenes are essentially hollow carbon shells of various sizes, the most well - known of these is a 60 - carbon unit called buckminster fullerene or C_{60} (more details see below). For instance, diamond is the hardest material, while graphite is one of the softest; diamond is transparent to the visible part of spectrum, while graphite is opaque; diamond is an electrical insulator, while graphite and graphene are a conductors. Very important is that all these different properties originate from the same carbon atoms, simply with different arrangements of the atomic structure. Below we describe the new phenomena of the carbon - isotope effect in diamond. Crystals ^{12}C and ^{13}C diamond differ only one neutron.

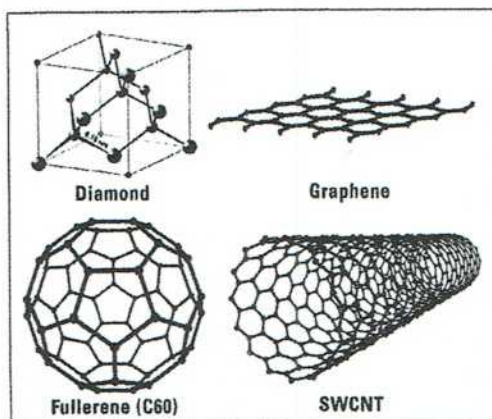


Fig. 7. Structure of some representative carbon allotropes [diamond, graphene, fullerene (C60) and SWCNT].

In the atomic ground state of carbon, the 6 electrons are in the configuration $1s^2 2s^2 2p^2$, i.e. 2 electrons fill the inner shell 1s, which is close to the nucleus and which is irrelevant for chemical reactions, whereas 4 electrons occupy the outer shell of 2s and 2p orbitals. Because the 2p orbitals ($2p_x$, $2p_y$ and $2p_z$) are roughly 4 eV

higher than the 2s orbital, it is energetically favorable to put 2 electrons in the 2s orbital and only 2 of them in the 2 p orbital (see, e.g. [1, 34]). It turns out, however, that in the presence of other atoms, such as e.g. H, O, or other C atoms, it is favorable to excite one electron from the 2s to the third 2p orbital, in order to form covalent bonds with the other atoms. The gain in energy from the covalent bond is indeed larger than the 4 eV invested in the electronic excitation. In the excitation state, we therefore have four equivalent quantum - mechanical states: $|2s\rangle$, $|2p_x\rangle$, $|2p_y\rangle$ and $|2p_z\rangle$. a quantum - mechanical superposition of the state $|2s\rangle$ and $n|2p_j\rangle$ states is called sp^n hybridization, which play an essential role in carbon bonds [34].

In the case of a superposition of the 2s and two 2p orbitals, which we may choose to be the $|2p_x\rangle$ and the $|2p_y\rangle$ states, one obtains the planar sp^2 hybridization. The three quantum - mechanical states are given by

$$|sp_1^2\rangle = \frac{1}{\sqrt{3}} |2s\rangle - \sqrt{\frac{2}{3}} |2p_y\rangle,$$

$$|sp_2^2\rangle = \frac{1}{\sqrt{3}} |2s\rangle + \sqrt{\frac{2}{3}} \left(\frac{\sqrt{3}}{2} |2p_x\rangle + \frac{1}{2} |2p_y\rangle \right), \quad (2)$$

$$|sp_3^2\rangle = -\frac{1}{\sqrt{3}} |2s\rangle + \sqrt{\frac{2}{3}} \left(-\frac{\sqrt{3}}{2} |2p_x\rangle + \frac{1}{2} |2p_y\rangle \right).$$

These orbitals are oriented in the xy - plane and have mutual 120° angles (Fig. 8^a). The remaining unhybridized $2p_z$ orbital is perpendicular to the plane. A prominent chemical example for such hybridization is the benzene molecule [35]. This molecule consists of a hexagon with carbon atoms at the corner linked by σ bonds (Fig 8^b). Each carbon atom has, furthermore, a covalent bond with one of the hydrogen atoms which stick out from the hexagon in a star like manner. In addition to the 6 bond, the remaining $2p_z$ orbitals form 3π bonds, and the resulting double bonds alternate

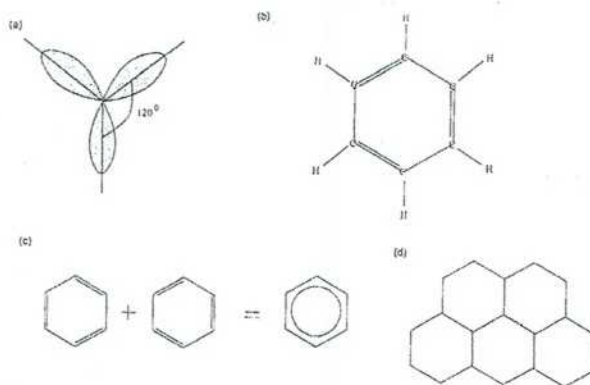


Fig. 8. (a) Schematic view of the sp^2 hybridization. The orbitals form angles of 120° . (b) Benzene molecule - C_6H_6 . The 6 carbon atoms are situated at the corners of a hexagon and form covalent bonds with the H atoms. In addition to the 6 covalent σ bonds between C atoms, there are three π bonds indicated by the doubled line. (c) The quantum - mechanical ground state of the benzene ring is a superposition of the two configurations which differ by the position of the π bonds. The π electrons are, thus, delocalized over the ring. (d) Graphene may be viewed as a tiling of benzene hexagons, where the H atoms are replaced by C atoms of neighbouring hexagons and where the π electrons are delocalized over the whole structure.

with single σ bonds around hexagon. Because a double bond is stronger than a single σ bond, one may expect that the hexagon is not perfect. A double bond ($C = C$) yields a carbon - carbon distance of 0.135 nm, whereas it is 0.147 nm for a single σ bond ($C - C$). However, the measured carbon - carbon distance in benzene is 0.142 nm for all bonds, which is roughly the average length of a single and a double bond. The ground state is a quantum - mechanical superposition of the two possible configurations for the double bonds shown schematically in Fig. 8^c. These chemical considerations indicate the way towards carbon - based condensed matter physics - any graphitic compound has been a sheet of graphene as its basic constituent. Such a graphene sheet may be viewed simply as a tiling of benzene hexagons, where one has

replaced the hydrogen by carbon atoms to form a neighboring carbon hexagon (see Fig 8^d). However, graphene has remained the basic constituent of graphitic systems during a long time only on the theoretical level. From experimental point of view, graphene is the youngest allotrope and accessible to physics measurements only since 2004. Graphite may be viewed as a stacking sheets (Fig.8^c) that stick together due to the van der Waals interaction, which is much weaker than in plane covalent bonds.

If one superimposes the 2s and all three 2p orbitals, one obtains the sp^3 hybridization, which consist of four club - like orbitals that mark a tetrahedron. The orbitals forma angles of 109.5° degrees (Fig 9^a). A chemical example for this hybridization is methane (CH_4), where the four hybridized orbitals are used to form covalent bonds with the 1s hydrogenatoms. In condensed matter physics, the $2p^3$ hybridization is at the origin of the formation of diamonds, when liquid carbon condenses under high pressure. The diamond lattice consists of two interpenetrating face - center - cubic (fcc) lattice as, with a lattice spacing of 0.357 nm as shown in Fig. 9^b.

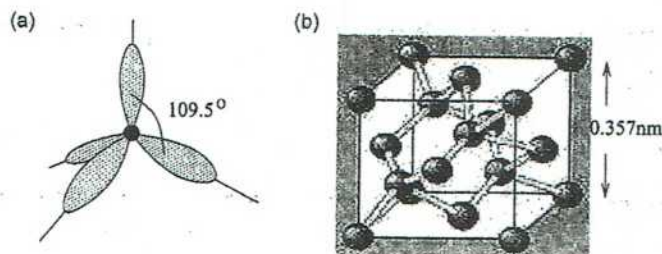


Fig. 9. (a) sp^3 hybridization with 109.5° angle between the four orbitals. (b) Crystal structure of diamond.

Although they consist of the same atomic ingredient, namely carbon, the 3D graphite and diamond crystals are physically extremely different. Graphite, as described above [1] is a very soft material due to its layered structure, whereas diamond is one of the hardest natural materials because all bonds are covalent σ bonds. The fact that all 4 valence electrons in the outer atomic shell are used in the

formation of the σ bonds is also the reason for diamond being an insulator with a large band gap of 5.47 eV (see above). In contrast to insulating diamond, the electrons in the weaker π bonds in graphite are delocalized and thus, yield good electronic conduction properties. The absorption spectrum demonstrating an indirect electronic transition in diamond is presented in Fig. 10.

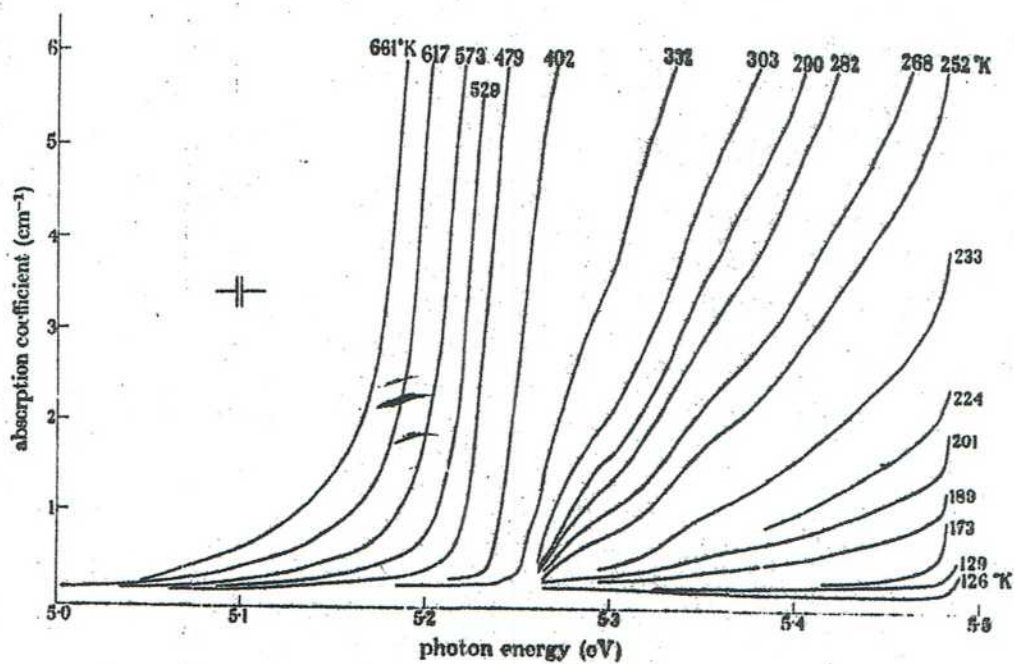


Fig. 10 Absorption edge spectrum of diamond at various temperature [36].

The indirect transition diagram is shown in Fig. 11. In diamond, we have seen (Fig. 2.5 in [37]) that excitation across the minimum separation of filled and empty states demands a large change in wave vector, and such a transition cannot be initiated by a photon unless it has access to a source of crystal momentum. We should repeat that it is the phonons that provide the required momentum (see, also Fig. 11).

We write the conservation laws in the form

$$\begin{aligned} E_f - E_i &= h\nu + \hbar\omega_f, \\ \vec{k}_f - \vec{k}_i &= 0 + \vec{q}, \end{aligned} \quad (3)$$

here $\vec{q}$ and ω_f apply to the phonon involved in the transition. Now $E_f - E_i = E_g$ and it is clear that the inclusion of phonons produces an absorption edge at a somewhat lower energy, namely $E_g - \hbar\omega_f$. These indirect or phonon - assisted transitions produce only weak absorption compared with that associated with direct transitions (see, e.g. [38]).

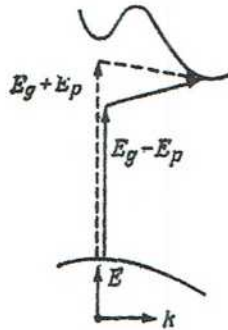


Fig. 11. Schematic of two - step transition in the case of the indirect bandgap of material, where E_p is the phonon energy and E_g is the energy of interband transition.

Due to the indirect gap of $E_g = 5.47 \pm 0.005$ eV (295 K), at $K = 0.76$ X, diamond has intrinsic phonon - assisted free exciton luminescence lines (see, e.g. [30]). The change of the indirect gap of diamond between pure ^{12}C and ^{13}C crystals has been determined by Collins et al. [39]. The luminescence spectra of the natural (^{12}C) and synthetic (^{13}C) diamond at electron excitation were investigated by Collins et al. [39] (Fig. 12), Ruf et al. [40], Watanabe et al. [41]. Fig. 12 compares the free exciton luminescence for a natural diamond with that for a synthetic diamond.

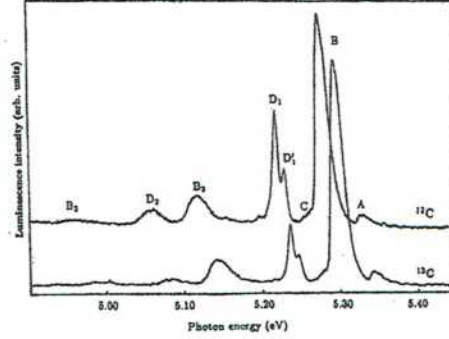


Fig. 12. Spectra measured at 77K phonon - assisted free cathodoluminescence feature (A, B and C) and the phonon assisted bound - exciton feature (D) from natural semiconducting ^{12}C diamond and a ^{13}C synthetic diamond (after [39]).

The peaks labeled A, B and C are due, respectively, to the recombination of a free exciton with the emission of transverse - acoustic, transverse - optic and longitudinal - optic phonons having wavevector $\pm k_{\min}$ and quanta in ^{12}C [30, 42].

$$\hbar\omega_{\text{TA}} = 87 \pm 2; \hbar\omega_{\text{TO}} = 141 \pm 2; \hbar\omega_{\text{LO}} = 163 \pm 1 \text{ meV.} \quad (4)$$

Features B_2 and B_3 are further free exciton processes involving the above TO phonon with one and two zone - center optic phonons respectively. As we can see from Fig. 12 the isotope shift of the free exciton luminescence spectrum of ^{13}C diamond is equal $16.5 \pm 2.5 \text{ meV}$ [30]. The more detailed and quantitative investigation of $E_g \sim f(x)$, where x is the isotope concentration was done by Ruf et al. [40] (Fig. 13) and Watanabe et al. [41] (Fig. 14) where five samples of diamond with different concentrations x were studied.

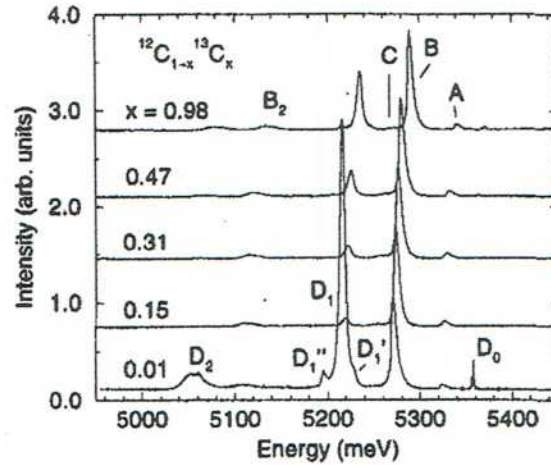


Fig. 13. Cathodoluminescence spectra of isotopically modified diamond at 36 K. Intrinsic phonon - assisted recombination peaks are labeled in the top spectrum, those from boron - bound excitons in that at the bottom. The spectra are normalized to the intensity of the B peak and vertically offset for clarity (after [40]).

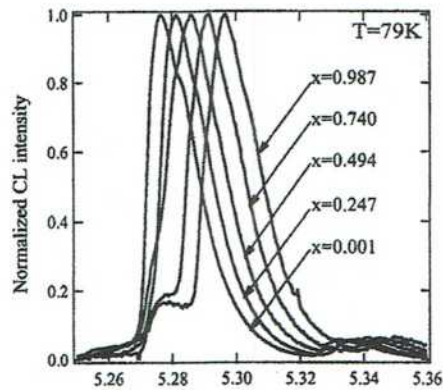


Fig. 14. luminescence spectra of free excitons in homoepitaxial diamond films

grown from mixture of methane in hydrogen by means of a microwave plasma - assisted CVD. The spectra illustrate the effect of isotope composition $^{12}\text{C}_{1-x}^{13}\text{C}_x$ mixed in the CVD gas phase. All spectra are normalized to the same height (after [41]).

Watanabe et al. have concluded that the maximum change of the band gap due to substitution of ^{12}C by ^{13}C is $\Delta E_g = 15.4$ meV. The dependence of the change exciton energy on the isotopic concentration in a diamond is depicted on the Fig. 15.

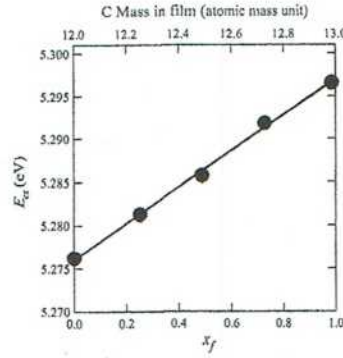


Fig. 15. Excitonic band - gap energy E_{exc} obtained for the CL peak energies in Fig. 14 as a function of the ^{13}C concentration of the films (x_f). The solid line is a linear regression fit of the data (after [41]).

This value (15.4 meV) is in good agreement with the estimate of 16.5 ± 2.5 meV by Collins et al. [36] using a zero - point renormalization obtained from a fit of the experimental temperature dependence of the band gap (see, also [39]). We should stress that the isotope shift in $^{12}\text{C}_x^{13}\text{C}_{1-x}$ diamond crystals approximately 15 meV per one neutron and on seven neutrons we get $15 \cdot 7 = 105$ meV. This value is very close to the observed one (0.103 meV) in $\text{LiH}_x\text{LiD}_{1-x}$ crystals. As will be shown below this value is the maximum value of the neutron - electron binding energy.

It is clear that the lattice - dynamic properties of a crystal are directly affected by the atomic mass. To a first approximation, phonon behave like harmonic oscillators

with frequencies [39]

$$\omega \propto m^{-1/2}. \quad (5)$$

Crystals containing various isotopes are usually described within virtual crystal approximation (VCA) [44] where m in Eq. (4) is replaced by the average atomic mass

$$\bar{m} = \sum_i c_i m_i. \quad (6)$$

It is obtained from the sum over the isotopes masses m_i and concentration c_i . In spite of VCA simplicity, the model describes the general feature of the lattice dynamics of mixed alkali - halide crystal sufficiently well (for details see [39, 42]). Isotopes are ideal for lattice - dynamic investigations of crystals. One can make use of the unique isotopic properties by varying the isotopic composition. Isotope substitution helps to disentangle the individual contributions of anharmonic and disorder - induced effects and to clarify the origin of phonon broadening mechanisms. Due to the fact that substitution of the isotopic mass in semiconducting crystals a small variation of E_g [42], perturbation theory is applicable (excluding LiH crystals [24]).

Raman spectroscopy is a powerful means to gain experimental access to phonons and their interaction and scattering mechanisms. All studies presented in this paragraph are restricted to stable, i.e. non - radioactive isotopes. About 300 stable and 1000 radioactive isotopes are known today. Some elements are isotopically pure (for example, Co), while others may contain numerous isotopic modifications (for example, Sn has 10 stable isotopes with atomic masses ranging from 112 to 124, while Xe has 23 isotopes, 9 of which are stable (see, Table 1 in [42])).

In this part, the modern understanding of first - order Raman slight scattering spectra in isotopically mixed elementary and compound (CuCl, GaN, GaAs, GaP) semiconductors having a zinc blende structure is described. It is well - known that materials having a diamond structure (C, Si, Ge, α - Sn) are characterized by the triply degenerate phonon states in the Γ - point of the Brillouin zone ($\vec{k} = 0$) (see, e.g. [37]). Isotope effect in light scattering spectra in Ge crystals was first investigated by Agekyan et al. [46]. A more detailed study of Raman light scattering spectra in

isotopically mixed Ge crystals has been performed by Cardona and coworkers [42].

The cubic modifications of crystalline carbon, diamond, is characterized by a tetrahedral coordination underlying its structure dictated by sp^3 bonding (see Fig. 9) between the nearest neighbor atoms. Diamond has two atoms per primitive (Bravais) cell. The strong covalent bonding and the light mass of the constituent atoms result in a large frequency (see, Eq. (3)) for zone center, Raman active, triply degenerate F_2 mode [47]. Its crystalline perfection and transparency [36] make diamond ideally suited for inelastic light scattering. First Raman study of the dependence the frequencies of phonons on the isotopic composition was conducted on diamond by Chrenko [49] in 1988. Several publications on diamond by other groups followed later [50 - 52]. Most clear results was obtained by Hanzawa et al. [48] (see Fig. 16).

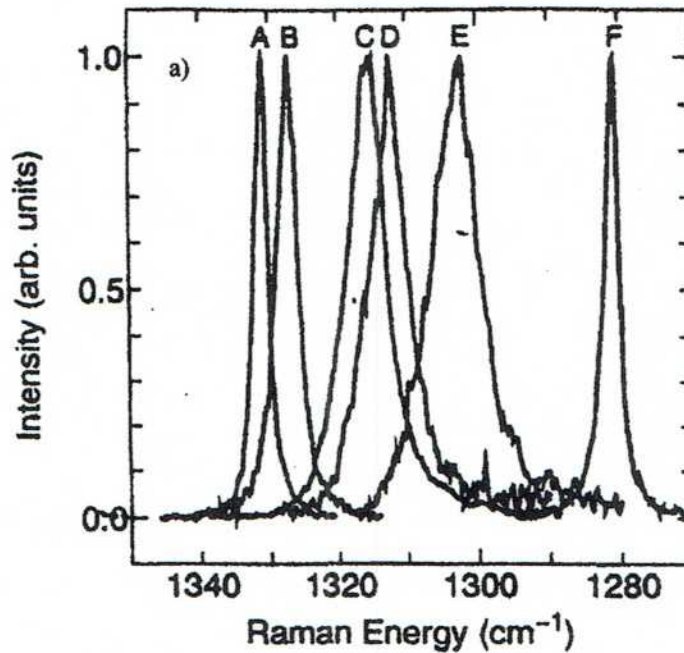


Fig. 16. First - order Raman scattering in isotopically mixed diamond crystals $^{12}\text{C}_x^{13}\text{C}_{1-x}$. The peaks A, B, C, D, E and F correspond to $x = 0.989; 0.90; 0.60; 0.50; 0.30$ and 0.001 (after [48]).

First - order Raman light scattering spectrum in diamond crystals also includes one line with maximum $\omega_{\text{LTO}}(\Gamma) = 1332.5 \text{ cm}^{-1}$ [51]. In Fig. 16 the first - order scattering spectrum in diamond crystals with different isotope concentrations is shown [48]. As was shown in [42], the maximum and width of the first - order scattering line in isotopically - mixed diamond crystals are nonlinearly dependent on the concentration of isotopes x . The maximum shift of this line is 52.3 cm^{-1} , corresponding to the two limiting values of $x = 0$ and $x = 1$. The effect of the isotopic ^{12}C to ^{13}C ratio on the first - and second - order Raman scattering of light in the diamond has been investigated in [52]. As ^{13}C content is increased from the natural ratio ($^{12}\text{C}/^{13}\text{C} = (1 - x)/x$, where $x = 0.011$ to the almost pure ^{13}C ($x = 0.987$) the whole spectrum has shifted towards longer wavelength (Fig. 17) in good agreement with the expected $M^{-0.5}$ frequency dependence on the reduced mass M . For an approximately equal mix of the two isotopes, the authors reported that the feature seen in the above two - phonon spectra were either broadened or unresolved.

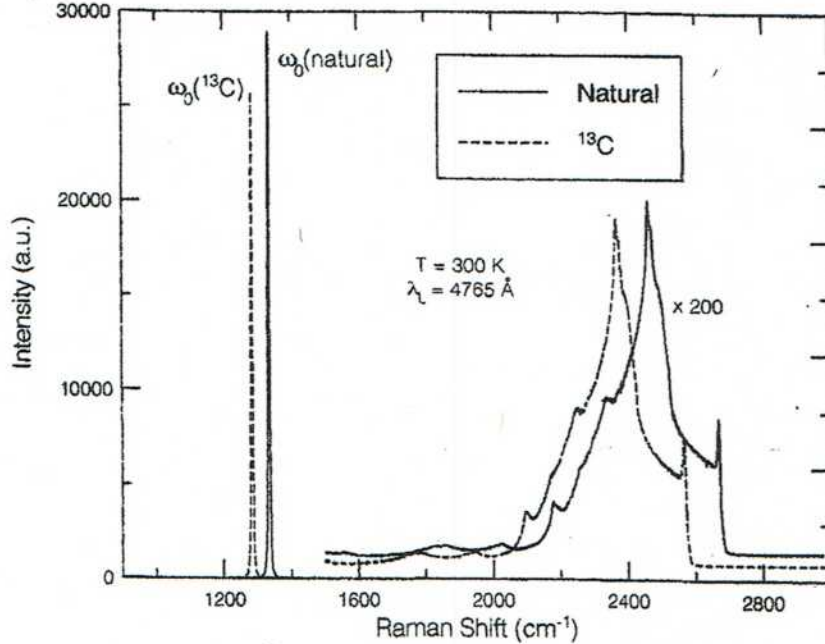


Fig. 17. The Raman spectra of natural and a ^{13}C diamond. The spectra show the dominant first - order Raman - active F_{2g} line and the significantly weaker quasi - continuous multiphonon features (after [52]).

The measured dependence on average mass mainly reflects the isotope effect on the harmonic frequency (Eq. (5)) which is, however, modified by anharmonic and disorder - induced contributions [30, 42]. Isotope renormalization frequencies of phonons of other semiconducting crystals (GaP, GaAs, α - Sn, ZnS, ZnO, CuCl, CuBr) was published in the next reviews [38]. We should highlight that addition one next neutron in nuclei is called the change in more or less of energy of elementary excitations in solids.

The science of the nuclear, atoms, simple molecules and the science of matter from microstructure to larger scales are well established. A remaining, extremely important [30, 42], size related challenge is at the atomic scale, roughly the dimensional scale between 1 and 10 molecular sizes, where the fundamental properties of materials are determined and can be engineered (see also [37]. This field of science - isotopetronics - is a broad and interdisciplinary field of emerging research and development. Isotopetronics is connected to materials, structures and systems which components, as in nanoscience, exhibit novel and significantly modified physical properties due to their small sizes [37]. The method of the isotope renormalization of the energy of elementary excitations in bulk solid and low - dimensional structures very often used in the last five decades and well documented in the scientific literature (see [30, 42] and references quoted therein). In this paper, current understanding of the band - gap opening in graphene is discussed along with associated experimental and theoretical investigations..

As was shown above, carbon, one of the second basic elements in nature after silicon, still gives a lot surprises. It is found in many different forms - allotropes - from zero dimensional fullerene, one dimensional carbon nanotubes, two dimensional graphene and graphite, to three dimensional diamond (Fig. 9) - and the properties of the various carbon allotropes can vary widely [20]. For instance, diamond is the hardest material, while graphite is one of the softest: diamond is transparent to the visible part of spectrum, while graphite is opaque; diamond is an electrical insulator, while graphite is a conductor. Very important is that all these different properties originate from the same carbon atoms, simply with different arrangements of the

atomic structure.

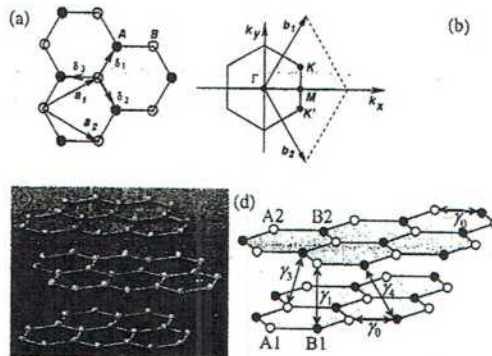


Fig.18. (a) Graphene honeycomb lattice showing the two triangular sublattices. (b) The graphene Brillouin zone in momentum space. (c) Lattice structure of graphite, graphene multilayer. (d) Lattice structure of bilayer graphene (explanation in text).

In two-dimensional graphene, carbon atoms are periodically arranged in an infinite honeycomb lattice Fig.18^a. Such an atomic structure is defined by two types of bonds within the sp^2 hybridization. From the four valence orbitals of the carbon atom (the $2s$, $2p_x$, $2p_y$, and $2p_z$ orbitals, where z is the direction perpendicular to the sheet), the (s , p_x , p_y) orbitals combine to form the in plane σ (bonding or occupied) and σ^* (antibonding or unoccupied) orbitals. Three σ -bonds join a C atom to its three neighbors. They are quite strong, leading to optical - phonon frequencies much higher than observed in diamond (see below). Such orbitals are even with respect to the planar symmetry. The σ bonds are strongly covalent bonds determining the energetic stability and the elastic properties of graphene. The remaining p_z orbital, pointing out of the graphene sheet is odd with respect to the planar symmetry and decoupled from the σ states. From the lateral interaction with neighboring p_z orbitals (called the $pp\pi$ interaction), localized π (bonding) and π^* (antibonding) orbitals are formed [54]. Graphite consists of a stack of many graphene layers. The unit cell in graphite can be primarily defined using two graphene layers translated from each other by a C-C

distance ($a_{c-c} = 1.42 \text{ \AA}$). The three-dimensional structure of graphite is maintained by the weak interlayer van der Waals interaction between π bonds of adjacent layers, which generate a weak but finite out-of-plane delocalization [35]. The bonding and antibonding σ bands are actually strongly separated in energy $> 12 \text{ eV}$ at Γ , and therefore their contribution to electronic properties is commonly disregarded, while the bonding and antibonding π states lie in the vicinity of the Fermi level (Fig. 19). The two remaining π bands completely describe the lowenergy electronic excitations in both graphene and graphite (see [35] and references therein). The bonding π and antibonding π^* orbitals produce valence and conduction bands (Fig. 19) which cross at the charge neutrality point (Fermi level of undoped graphene) at vertices of the hexagonal Brillouin zone. Carbon atoms in a graphene plane are located at the vertices of a hexagonal lattice.

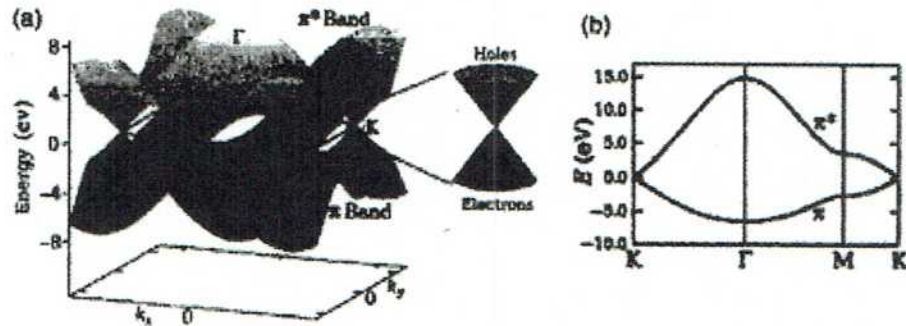


Fig. 19. Energy dispersion of graphene obtained within the tight - binding approximation. a) Energy dispersion relation for graphene, drawn in the entire region of the Brillouin zone. Since in this approximation to ignore the coupling between the graphene sheets, the bands depend only on k_x and k_y . The π band is completely filled and meets the totally empty π^* band at the K points. Near these points both bands linear dispersion as described in the literature. b) The dispersion along the high symmetry points Γ MK.

This graphene network can be regarded as a triangular Bravais lattice with two

atoms per unit cell (A and B). Each A- or B - type atom is surrounded by three atoms of the opposite type. In a simple neighbor model graphene is a semimetal with zero - overlap between valence and conduction bands. The energy dispersion of π electrons in graphene was first derived in 1947 by Wallace [54] within the tight - binding approximation. In this case, the wave function of graphene is a linear combination of Bloch function for sublattice A

$$\Phi_A = \frac{1}{\sqrt{N}} \sum_{\vec{R}_A} e^{i\vec{k}\vec{R}_A} \varphi(\vec{r} - \vec{R}_A), \quad (7)$$

and equilibrium function Φ_B for the B sublattice. Here N is the number of unit cells, $\vec{R}_A$ are the position of the atom A and $\varphi(\vec{r} - \vec{R}_A)$ is the $2p_z$ orbital of the atom at $\vec{R}_A$. The sum runs over all unit cells, i.e. all possible lattice vectors. In the nearest neighbor approximation (every A site has three nearest B sites, and vice versa), the energy eigenvalues can be obtained in a closed form [35]

$$\varepsilon(k_x, k_y) = \pm \gamma_0 \left[1 + 4\cos\frac{\sqrt{3}k_x a}{2} \cos\frac{k_y a}{2} + 4\cos^2\frac{k_y a}{2} \right]^{1/2}, \quad (8)$$

where γ_0 is the transfer integral between the nearest neighbors. The energy dispersion of two - dimensional graphene according to this formula is plotted in Fig. 2(a) as a function of the wave vector $\vec{k}$. The upper half of the curves is called the π^* or the antibonding band while the lower one is π or the bonding band. The two bands degenerate at the two K points given by the reciprocal space vectors $\vec{K} = (2\pi/a)(1/3, 1/\sqrt{3})$ and $\vec{K}' = (2\pi/a)(-1/3, 1/\sqrt{3})$ points where the dispersion vanishes (see above).

Basically, graphene has redefined the limits of what a material can do: it boasts record thermal conductivity and the highest current density at room temperature ever measured (a million times that of copper!); it is the strongest material known (a hundred times stronger than steel!) yet is highly mechanically flexible; it is the least permeable material known (not even helium atoms can pass through it!); the best transparent conductive film; the thinnest material known; and the list goes on ...[34]. In the vicinity of K - points (as it can be see from Fig. 19), the low - energy electron/hole dispersion relation is proportional to momentum, rather than its square. This is analogous to the energy dispersion relation of massless relativistic electrons, so

the electrons/holes of graphene are described as Dirac fermions having no mass. In a simple neighbor model graphene is a semimetal with zero - overlap between valence and conduction bands. In order to make graphene a real technology, a special issue must be solved: creating an energy gap at K - points in the Brillouin zone [35]. Different attempts have been made by researches, such as patterning graphene into nanoribbon, forming graphene quantum dots], making use of multilayer graphene sheets and applying an external electric field [55 - 58] It was shown that the uniaxial strain can open a band - gap in a metallic carbon nanotubes as well as carbon nanoribbon [59 - 61].

As it can be seen from Fig. 12 the band gap of ^{13}C has increased by 13.6 meV. Numerous examples of band gap increasment at hard isotope substitution were collected in the papers [30, 42]. The effect of the isotopic ^{12}C to ^{13}C ratio on the first and second - order Raman scattering of light in the diamond has been investigated in [41, 52]. As the ^{13}C content is increased from the natural ratio ($^{12}\text{C}/^{13}\text{C} = (1 - x) / x$, where $x = 0.011$), to the almost pure ^{13}C ($x = 0.987$), the whole spectrum has shifted towards longer wavelengths (see Fig. 16) in good agreement with the expected $M^{-0.5}$ frequency dependence on the reduced mass M . For an approximately equal mix of the two isotopes, the authors reported that the features seen in the above two - phonon spectra were either broadened or unresolvable. We should stress that the main line in the first - order Raman scattering spectrum of light at ω 1332 cm^{-1} also shifts to the short-wavelength side on the 52.3 cm^{-1} [30, 42].

Elastic and inelastic light scattering are powerful tools for investigating graphene [62 - 67]. Raman spectroscopy allows monitoring of doping, defects, disorder, chemical and isotope [1] modifications, as well as edges and uniaxial strain (see Fig. 20).

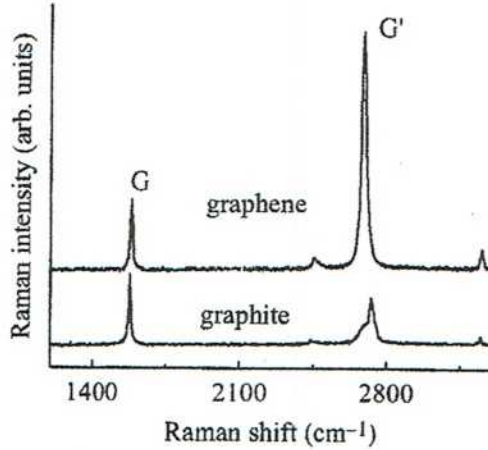


Fig. 20. Comparison of the Raman spectra of graphene and graphite measured at room temperature and at the excitation at $E_{laser} = 2.41$ eV (514.5 nm).

All sp^2 - bonded carbons show common features in their Raman spectra, the so - called G and D peaks (see, Fig. 20), around 1580 and 1360 cm^{-1} (see, e.g. [34]). The G peak (see, also below Fig. 22) corresponds to the E_{2g} phonon at the Brillouin zone center (Γ - point). The D peak is due to the breathing modes of six - atom rings and requires a defect for its activation. It comes from TO phonons around the Brillouin zone K point and it is activated by an intravalley scattering process. The 2D peak is the second order of the D peak. This is a single peak in monolayer graphene, whereas it splits into four bands in bilayer graphene, reflecting the evolution of the band structure [1, 34]. The Raman spectrum of graphene also shows significantly less intensive defect - activated peaks such as the D' peak, which lies at ~ 1620 cm^{-1} . This is activated by an intravalley process i.e. connecting two points belonging to the same cone around K (see, Fig. 6 in [1]). The second order of the D' peak is called 2D' peak. Since 2D and 2D' peaks originate from a Raman scattering process where momentum conservation is obtained by the participation of two phonons with opposite wave vector ($\vec{q}$ and $-\vec{q}$), they do not require the presence of defects. Thus, they are always visible in the Raman spectrum (see cited papers [19 - 25] and references therein).

Graphene is one unique material which shows properties not found in other

materials. One of these unique features of graphene is the influence of long range strains on the electronic properties. The possibility of tuning the dynamics of carriers as well as phonons by appropriately designed strain patterns opens the way for novel applications of graphene, not possible with any other materials (see, e.g. [25] and references therein). At present time we have several reports, which have examined graphene properties under uniaxial deformation [1, 34, 68, 69].

Strain can be very efficiently studied by Raman spectroscopy since this modifies the crystal phonon frequency, depending on the anharmonicity of the interatomic potentials of the atoms. Raman spectra of strained graphene show significant redshifts of 2D and G band (see Table 2) because of the elongated of the carbon - carbon bonds.

Table 2. Red shift of the G and 2D bands in the Raman spectra in graphene monolayers under uniaxial tensile stress.

Ref.	Shift of G (G^+ and G^-) band $\text{cm}^{-1}/\%$	Shift of 2D band $\text{cm}^{-1}/\%$	E_g , meV
15	14.2	27.8	300
25	5.6; 12.5	21	
27	10.8; 31.7	64	
26			≈ 500
theory			

The authors of the paper [68] have proposed that by applying uniaxial strain on graphene, tunable band - gap at K - point can be realized. First principle calculations predicted a band - gap opening of ≈ 300 meV for graphene under 1% uniaxial tensile strain (Fig. 21). Thus, the strained graphene provides an alternative way to experimentally tune the band - gap of graphene, which would be more efficient and more controllable than other methods (see, above) that are used to open band - gap in graphene.

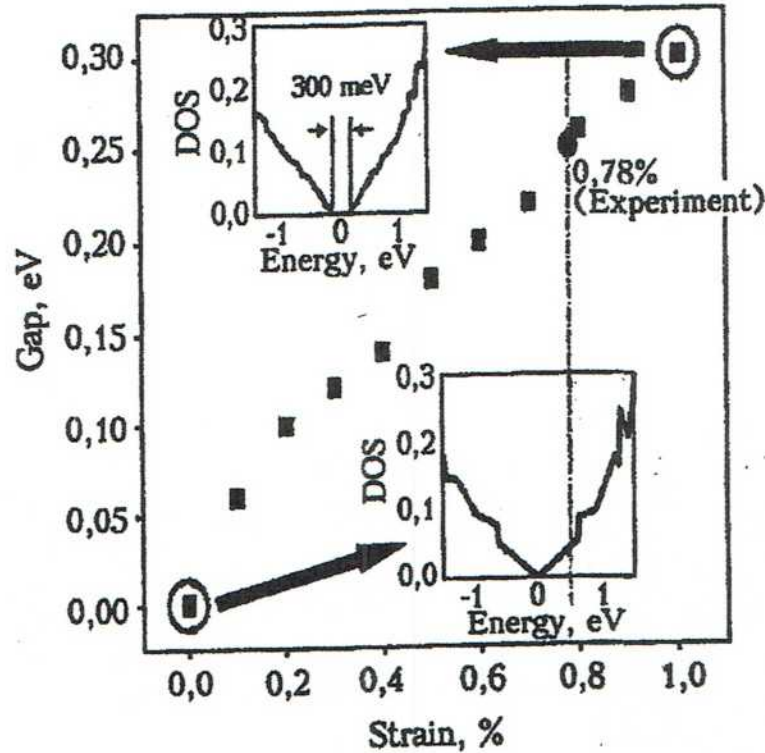


Fig. 21. The band - gap of strained graphene with the increase of uniaxial tensile strain on graphene. The magnitude of gap is determined by the gap opening of density of states. The insert show the calculated density of states of unstrained and 1% tensile strained graphene. The dash line and solid dot indicate the calculated bandgap of graphene under the highest strain (0.78 %) [68].

The method of the isotope renormalization of the energy of elementary excitations in solid very often used in last five decades and well described in the scientific literature (see, for example reviews [24, 30, 42]). At now there is a large list of the paper devoted to investigation of the isotope - mixed graphene [1, 34, 70]. Chen et al. [65] have reported the first experimental study of the isotope effect on the thermal properties of graphene. The thermal conductivity K , of isotopically pure ^{12}C (0.01 of ^{13}C) graphene determined was higher than 4000 W/mK (approximately two times

more than it in diamond [41, 51]) at the measured temperature $T_m \sim 320\text{K}$, and more than a factor of two higher than the value of K in a graphene sheets composed of a 50% - 50% mixture of ^{12}C and ^{13}C . Raman spectroscopy transferred to the 285 nm SiO_2/Si wafer was performed under 532 nm laser excitation [65]. The G peak and 2D band positions in Raman spectra of graphene with 0.01%, 1.1%, 50% and 99.2% ^{13}C - isotope are presented in Fig. 22.

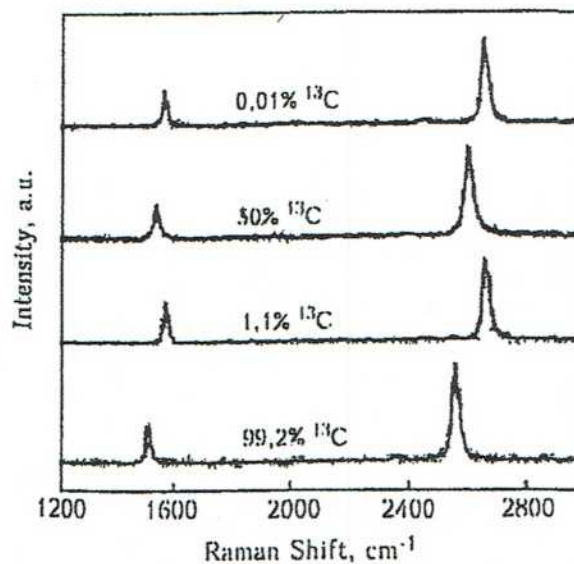


Fig. 22. Raman spectra of graphene with different isotope concentration at room temperature [65].

Isotope shift of the G and 2D bands in the Raman spectra depicted on the Fig. 23 [33].

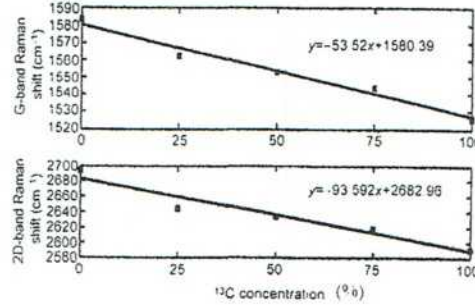


Fig. 23. Peak position of G and 2D bands in Raman spectra as a function of the concentration of ^{13}C [70].

As in the case of isotope - mixed diamond [30, 34, 42] the Brillouin - zone - center optical - phonon frequency ω varies with the atomic mass M as $\omega \sim M^{-1/2}$ making the Raman shift for ^{13}C approximately $(12/13)^{-1/2}$ times smaller than that for ^{12}C . The experimental difference between the lowest 99.2% ^{13}C and the highest 0.01% ^{13}C peak is $\sim 64 \text{ cm}^{-1}$ which is according [42] in agreement with the theory, and attests for the high quality of isotopically modified graphene. By the way we should indicate that in the Raman spectra in diamond (with sp^3 - bond) analogous shift of first - order line in Raman spectrum is equal 52.3 cm^{-1} [34], which is consistent with the isotope mass ratio. Substituting a light isotope (^{12}C , H) with a heavy one increases the interband transition energy in the case $^{12}\text{C}_x^{13}\text{C}_{1-x}$ on 14.7 meV and $\text{LiH}_x\text{D}_{1-x}$ on 103 meV [24]. Taking into account a more soft bond (sp^2 - bond instead sp^3 - bond in diamond) isotope - induced band - gap opening in graphene of some hundreds meV (up to E_g of Si) was predicted in paper [71]. Such estimation of the value of isotopical band - gap opening in graphene agrees with not only the results of paper [34] but with very small value $C_{44} = 0.5 \cdot 10^{10} \text{ dyn/cm}^2$. Such small value indicates on the strong electron - photon interaction - main reason renormalization of electron excitation energy (for the details, see, e.g. [1]). Very close to isotopically renormalization of electronic excitation energy is the hydrogenation of graphene [71]. In last mechanism there is observable band - gap opening in graphene. We should add that use deuterium instead of hydrogen we may increase the value of E_g [30, 42]. Thus, isotope substitution will be very useful method for renormalization of the band - gap in graphene - future semiconducting material. Moreover, this method allows to control not only of the

strong nuclear interaction (quantum chromodynamics) but taking it into account at the renormalization of the electromagnetic interaction (quantum electrodynamics) [1]. Adding ^{13}C makes magnetic materials isotope out of graphene.

IV. Discussion.

Traditionally nuclear - electron interaction (in our case neutron - electron interaction) taking into account the solving of Schrödinger equation using Born - Oppenheimer (adiabatic) approximation [72]. Since electrons are much faster and lighter than the nuclei by a factor nearly 2000 the electron charge can quickly rearrange itself in response to the slower motion of the nuclei, and this is the essence of the Born - Oppenheimer approximation. This approximation results the omission of certain small terms which result from the transformation. As was shown in [30] the eigenvalue (energy) of the electronic Schrödinger equation (equation 6 in [73]) depends on the nuclear charges through the Coulomb potential, but it doesn't include any references to nuclear mass and it is the same for the different isotopes. The independent of the potential energy (the eigenvalue of the Schrödinger equation) is the essence adiabatic approximation. However, we must repeat, that the Born - Oppenheimer approximation is the standard ansatz to the description of the interaction between electrons and nuclei in solids (see, e.g. [74]). The last result is forcing us to search for new models and mechanisms of nuclear - electron interaction including results of subatomic physics, e.g. hadron - lepton interaction.

Out of four known interactions, three are described by SM - the electromagnetic, weak and strong ones [1 - 5]. The first two of them have a common electroweak gauge interaction behind them. The symmetry of this interaction $SU(2)_L \times U(1)_Y$ manifests itself at energies higher than ~ 200 GeV. At lower energies, this symmetry is broken down to $U(1)_{EM} \neq U(1)_Y$ (the electroweak symmetry breaking): in SM this breaking is related to the vacuum expectation value of a scalar field [75]. The strong interaction in SM is described by the QCD, a theory with the gauge group $SU(3)_C$. The effective coupling constant of this theory grows when the energy decreased. As a result, particles which feel this interaction cannot exist as free states and appear only in the form of bound states called hadrons (see, also [1]). Most of modern methods of quantum field theory work at small values of coupling constant, α_s , [75], that is, for

QCD, at high energies. Quarks and leptons, the so - called SM matter fields, are described by fermionic fields. Quarks take part in strong interactions and compose observable bound state hadrons. Both quarks and leptons participate in the electroweak interaction. The matter fields constitute three generation: particles from different generation interact identically but have different masses (see, e.g. [4]). For the case of neutrino, Yukawa interactions are forbidden as well, so neutrinos are strictly massless in SM (see, however [73] and references therein). The gauge bosons, which are carriers of interactions, are massless for unbroken gauge groups $U(1)_{EM}$ (electromagnetism - photons) and $SU(3)_C$ (QCD - gluons), masses of $W^\pm$ and Z_0 bosons are determined by the mechanism of electroweak symmetry breaking. It should be noted that the forces between the quarks must be long range, because the gluons have zero mass. This does not imply that forces between hadrons are also long range, because hadrons have zero color charges overall. The forces between the colorless hadrons are the residues of the forces between their quark constituents, and cancel when the hadrons are far apart.

Returning to our non - accelerator experimental results, we should underline that in this paper we measure the strong nuclear interaction in crystals which differ by term of one neutron from each other. When we add one neutron in the hydrogen nucleus, we artificially activate the strong interaction. As far as the gravitation, electromagnetic and weak interactions are the same in both kind crystals (LiH and LiD), it only changes the strong interaction. Therefore a logical conclusion is made that the renormalization of the energy of electromagnetic excitations (isotopic shift equals 0.103 eV) is carried out by strong nuclear interaction. The short range character of the strong interaction of nucleons does not possess direct mechanism of the elementary excitation (electrons, excitons, phonons) energy renormalization, which was observed in our low temperature experiments. Second reason that the interpretation of our experimental results is a very difficult task because they are the first demonstration of the violation of the strong conclusion in nuclear and particle physics that the strong nuclear force does not act on the colorless leptons (see, e.g. [1, 73]). Moreover we have some contradictions taking into account that the forces between quarks must be long range, because the gluons have zero mass. But as was mentioned above, the short range when forces between the colorless hadrons are the residues of the forces between their quark constituents, and cancel when the distance between hadrons is more than nuclear size [5]. We can see that the nuclear size transforms long range interaction in the short range strong one. It is a very old question which up to present time has not any theoretical explanation.

In spite of above discussion, at present time we can distinguish the following

mechanisms of the isotopic shift zero phonon line in LiD crystals:

1. Long range electric field of the neutron's quarks. This mechanism owing to the confinement quarks is limited by the boundary of the neutron.

2. The possible new structure of the quarks and leptons - so - called preons (see e.g. [75] and references quoted therein).

3. The most likely mechanism of the neutron - lepton interaction is connected to the magnetic - like strong field of neutron's quarks. Taking into account anomalous magnetic moment of the neutron [14] in the paper [6] was obtained the value of strong coupling constant $\alpha_s = 2.4680$. Quite large value in comparison with the accelerator technique value $\alpha_s(M_Z) = 0.1198$ [76]. The large value α_s is thus justified to think that residual strong forces acting beyond nucleon could exist. Besides that our experimental results to allow the neutron electron binding energy equal 106 meV was found in diamond and LiH which is 128 times less than proton - electron coupling energy (13.6 eV) and its value is perfectly agree with Breit's theory. The results of our experiments make it possible to determine not only the constant of strong nuclear interaction equal 2.4680, but also to predict the mass of the boson carrying the interaction between neutron and electron equal 3.5 keV [21].

An another possible interpretation is to assume that in addition to the 8 gluons predicted by QCD $SU(3)_c$ group there is a ninth gluon color singlet [4]

$$g_9 = \frac{1}{\sqrt{3}} (\bar{r}r + \bar{g}g + \bar{b}b) \quad (9)$$

This massless photon - like gluon may be strongly interacts between nucleons (neutrons) and leptons (electrons) (see, also [1]). Returning to Fig. 6 we can note that our measurements permit value obtainment of strong coupling constant from $\alpha_s = 2.4680$ (pure LiD crystals) to $\alpha_s = 0$ (pure LiH crystals).

The remarkable properties of the graphene promise unheard of applications, especially in the semiconductor state. Especially graphene nanostructures are promising candidates for future nanoelectronics and solid - state quantum information technology. As was shown numerously, in the vicinity of K - points (see, Fig. 6 in [1]), the low - energy electron/hole dispersion relation is proportional to momentum, rather than its square (see, e.g. [78]). This is analogous to the energy dispersion relation of massless relativistic electrons, so the electrons/holes of graphene are described as Dirac fermions having no mass. In a simple neighbor model graphene is a semimetal with zero - overlap between valence and conduction bands. In order to make graphene a real technology, a special issue must be solved: creating an energy gap at K - points in the Brillouin zone (see, Fig. 24).

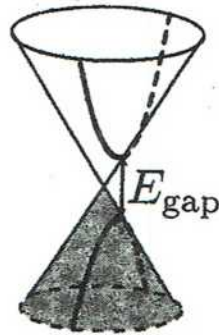


Fig. 24. The energy dispersion of semimetal (solid line) and semiconductor (dashed line).

Different attempts have been made by researches, such as patterning graphene into nanoribbon, forming graphene quantum dots, making use of multilayer graphene sheet and applying an external electric field [79]. It was shown that the uniaxial strain can open a band - gap in a metallic carbon nanotubes as well as carbon nanoribbon . As an example Fig. 25 shows the dependence of the energy of interband transition on the width of the graphene nanoribbons. The observed dependence is well described by the following expression $E_g = \alpha^* / (W - W^*)$, where $\alpha^* = 0.38$ eV/nm, and $W^* = 16$ nm accounts for inactive edges (see [80] and references quoted therein). This formula gives value of E_g equal to several eV, if W is reduced to a size , for example, ^{13}C , but it is impossible. The isotopic transformation of graphene, a semimetal into semiconductor deserves special attention. This means of the cones dispersion pattern $E(\vec{k})$ at the K point of the Brillouin zone of graphene reformes into the usual parabolic dispersion of band structure pattern (see Fig. 24).

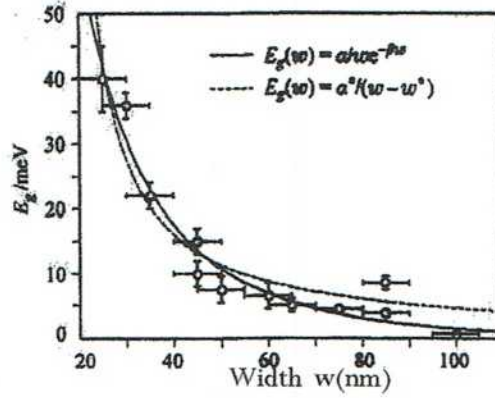


Fig. 25. The dependence of E_g as a function of width of the graphene nanoribbons (after [80]).

According to the results of Ref. 79 this method can be used to obtain E_g of several hundred meV. Since value of E_g depends on the concentration of ^{13}C in graphene, this mechanism is easily realized either chemically or technologically. With the opening E_g of massless fermions (leptons = electrons) acquire mass. estimates of different ways of opening E_g in graphene give the following equality: $E_g = 2m$ [78]. When $E_g = 1$ eV [71] leptons acquire a mass equal 0.5 eV - the value is not large, but finite. One of the mechanisms of acquire mass in elementary particle physics is considered in the paper [81]. This paper predicts that both quarks and gluons acquire running mass distribution in QCD, which are large at infrared momenta. The current quark of perturbative QCD evolves into a constituent quark as its momentum becomes smaller. The constituent quark mass arises from a cloud of low - momentum gluons attaching themselves to the current quark. This is DCSB [82]: an essentially nonperturbative effect that generates a quark mass from nothing; namely it occurs even in the chiral limit. DCSB namely the generation of mass from nothing is a theoretically established nonperturbative feature of QCD. The solution of a gap equation shows that gluons are cannibals: they are particle species whose members become massive by eating each other (the details see [79, 81, 82]).

Thus, the tentative interpretation of describing non - accelerator experimental results does not find consistent explanation at the change of strong interaction leaving to another mystery of SM [3]. We should remind that intrinsic contradiction of Standard Model is already well - known. Really, the Lagrangian of quantum chromodynamics (theory of the strong interaction) has the next form (see, e.g. [2 - 4]):

$$L = i \sum_q \bar{\Psi}_q^a (\nabla_\mu \gamma_\mu + im_q) \Psi_q^a - \frac{1}{4} G_{\mu\nu}^n G_{\mu\nu}^n, \quad (10)$$

where

$$\begin{aligned} \nabla_\mu &= \partial_\mu - ig \frac{\lambda^n}{2} A_\mu^n, \\ G_{\mu\nu}^n &= \partial_\mu A_\nu^n - \partial_\nu A_\mu^n + gf^{nml} A_\mu^n A_\nu^l. \end{aligned} \quad (11)$$

Ψ_q^a and A_μ^n are quark and gluon fields, $a=1,2,3,\dots,8$ are color indices, λ^n and f^{nml} are Gell - Mann matrices and f symbols, m_q - are bare (current) masses, $q = u, d, s, c, \dots$ different quarks. It is common place [1, 76] that the Lagrangian (10) contains the members which describe both free motion and interaction between quarks and gluons, which is defined by the strength couple g . Spacing of which it is necessary to remark that although the Lagrangian (10) possesses rather attractive peculiarities (see, also [1, 2]), its eigenstates are the quarks and the gluons which are not observed in free states [79, 83]. The observed hadrons in the experiment don't eigenstates in quantum chromodynamics. It is obvious to expect that the modern theory of QCD should finally overcome these difficulties, for example inserting QCD in QED.

V. Conclusion.

The artificial activation of the strong nuclear interaction by adding one (two or more) neutrons in atomic nuclei leads it to the direct observation of the long - range strong interaction in low - temperature optical spectra of solids. This conclusion opens new avenue in the investigation of the constant of strong nuclear interaction in the wide value range by means the condensed matter alike traditional methods. Our experimental nonaccelerator results may shed light on a number of fundamental puzzles in modern physics, particularly on the unification of forces. Experimental observation of the renormalization of the elementary excitation energy of solids by the

strong nuclear interaction stimulates its count in the process of description of the elementary excitations dynamics in quantum electrodynamics. Besides, we should highlight that such important information has been obtained via rather simple and inexpensive experimental physics equipment. Present article continuous to develop between nuclear, high energy and condensed matter physics taking into account that our result does not available on accelerators.

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